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Concomitance in single bubble sonoluminescence of period doubling in emission and shape distortion

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ABSTRACT

We report the first direct observation for a single stable sonoluminescing bubble of a shape instability. Furthermore we show that stable saturation of the shape distortion caused by the instability for a certain range of parameters is experimentally possible and furthermore is directly linked to the curious phenomenon of flash by flash period doubling of the sonoluminescent emission as the afterbounce instability causing the shape distortion is always period doubled whenever the emission is & vice versa.

1. Introduction

In single bubble sonoluminescence (SBSL) a resonant ultrasound field drives a bubble repeatedly into a dramatic collapse, so violent as to heat the bubble sufficiently for light to be emitted in a pico-second long flash once per period of the sound. Basically the bubble dynamics is well understood to the point where we even know the mechanisms that lead to annihilation of the bubble (for a review see e.g. [1]). Several shape instabilities exist, described theoretically by expansion in spherical harmonics [2–5], and experimental studies [6,7] showed close coincidence between the theoretically determined onset of shape instabilities of order $n = 2$ or 3 (referring to the relevant spherical harmonics) and the destruction of the bubble. For diagrams over the relevant R_0, P_A space and the experimental confirmation we refer the reader to e.g. Refs. [6–8]. Here R_0 and P_A are respectively the ambient bubble size and the applied ultrasound amplitude. However, none of these instabilities have ever been observed directly for SBSL in a situation where light is emitted, although instabilities were observed below threshold for the emission by Matula [9] and for a bubble that emitted light before but not after an instability occurred [10]. Finally we should add that the different types of instabilities were cataloged by Hilgenfeldt et al. [3] as a Rayleigh–Taylor type instability and an afterbounce driven parametric resonance that both could be triggered by noise and occurred within a single period of the drive and a *pure* parametric instability driven by the drive and thus acting over several periods.

With the relative concentration of argon used in the present experiment, the shape instability of interest here is that caused by the parametric resonance which is driven by the afterbounces. As often the case where parametric amplification is present, the result is an oscillation with twice the periodicity of the driving oscillation, i.e. here the afterbounce oscillation.

A different issue is the occurrence of spatially anisotropic emission [11]. Given the right conditions this *spatially anisotropic emission* [12,13] may even period double (PD) without affecting the strict periodicity of the timing. This is in striking contrast to earlier findings by Holt et al. [14] of a period doubling cascade to chaos in the *timing of pulses*. None of these phenomena has ever found a satisfactory explanation. For instance, though Parlitz et al. [15] and Simon et al. [16] both found that period doubling could indeed be present in the bubble dynamics without breaking the spatial symmetry, the relevant parameter regime was far beyond the regime of sonoluminescence. Recently Holzfuss [17] suggested on the basis of simulations that an observed PD in the afterbounce spatial instability could be connected to the source of that in the emission, so far the best explanation for this phenomenon. The merits and shortcomings of this idea will be discussed in Section 5. Excitingly, linking the two phenomena gives additional credit to the picture of a hot core producing most of the light in combination with refraction in a distorted bubble surface as also suggested by the associated spatial symmetry breaking [13,18].

Finally we would like to point out that a better understanding of the issues raised here is of interest in the fields of sonoporation and

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sonophoresis, see e.g. Refs. [19–21], and inertial confinement fusion see e.g. Ref. [22] where similar shape instabilities conceivably may be observable.

2. Experimental setup

One of the difficulties relating to the measurement of possible shape oscillations for SBSL is the sub-micron size at the time of emission. The bubble is too small to resolve by imaging. To overcome this we have used Mie scattering [23] (see e.g. Ref. [24] for details when used for scattering from bubbles). To ensure that deviations from the signal expected for a sphere are real, we use two detectors. The scattered laser light inside a cone of solid angle $\Omega \approx .2$ is focused by a lens onto a 1 mm diameter pin hole and detected by a Hamamatsu R6357 photomultiplier (PMT) chosen for its high sensitivity at the laser wavelength of 632 nm. Two such systems are placed in the horizontal plane of the bubble so they detect the forward scattered light centered around an approximate angle of 70°. To a very high precision, the use of this geometry results in the scattered intensity being proportional to the bubble radius squared although what is actually measured at the exact time of the collapse is uncertain due to the strong compression of the surrounding water and the emission of sonic shock waves. The sensitivity is sufficiently high for us to follow the bubble oscillations live without having to resort to averaging. A similar detection system is used for the emission in order to prevent laser light scattered or reflected from the surroundings from reaching that detector. Here, though, the Hamamatsu R6354 solar blind PMT is chosen in order to further exclude the laser light. Again, the sensitivity level is high enough to observe any substantial PD directly on an oscilloscope. The undisturbed part of the laser beam passes to a beam dump to minimize noise due to reflections. A sketch of the complete system is shown in Fig. 1. The system is placed in a lightproof enclosure and cooled to the operating temperature (usually 9 °C).

The preamplified PMT signals are fed to a National Instrument acquisition system consisting of a NI PXIe-1062Q chassis with a PXIe-8133 embedded controller and two PXIe-5122 100 MHz, 14 bit digitizers. Data are streamed to a NI HDD-8264 memory-bank using a PXIe-8262 rate controller. The system allows for 4 channel, 100 MHz simultaneous recording for about 2 s, and 'unlimited' when run at 50 MHz. Files are, however, limited to approximately 3 GB. NI DIAdem is used for preliminary analysis while the final analysis is done with MATLAB.

The vessel used is of the same design as in previous work and consists of a quartz cylinder, 6 cm in height and 6 cm in diameter, with steel caps at both ends [13]. The resonator is sealed using a pressure relief bag. Piezoelectric transducers are mounted on both caps for the drive, which has a frequency of approximately 22,000 Hz at room temperature. The drive signal is produced by an Agilent 33220A function generator, amplified by an ILP HY2005 power amplifier and fed to the transducers through a home-made tuning circuit. Air is dissolved in the water at a reduced pressure of about 400 mbars referred to room temperature. From experience we know that PD is present in this system [13]. At the operating temperature of 9 °C for the experiment, the relative argon concentration and the resonance frequency are approximately 0.0025 and 21,800 Hz respectively. The set-up of the cell with the surrounding optics as placed in the cooled enclosure is depicted in Fig. 2 with the numbers, indicating the different elements of the set-up, explained in the figure caption. The 20 mW helium–neon laser used for the Mie-scattering is placed outside the enclosure with the laser beam fed in through a 3 mm pin-hole.

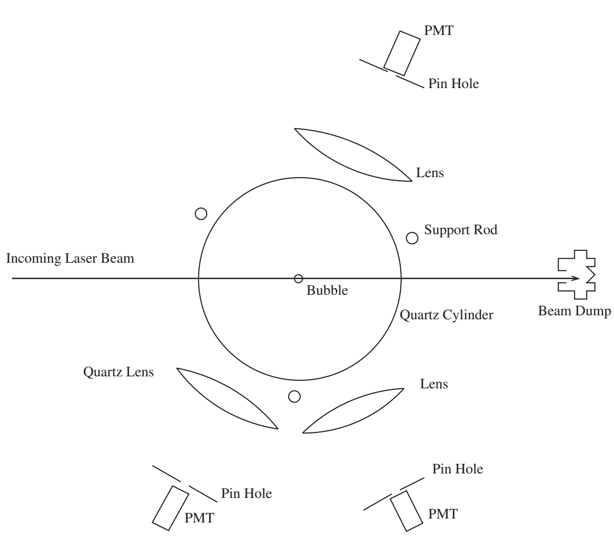

Fig. 1. Outlay of the detection scheme (not to scale) in the horizontal plane including the bubble. The position of the laser beam (slightly slanted with respect to the plane to avoid hitting the bubble with a reflection) and one of the Mie scattering detectors are externally controlled. The constraints imposed by the support rods for the cylindrical vessel unfortunately do not allow for a fourth detection system, nor is there space in the lightproof enclosure for external control of the other two systems resulting in some minor degradation of signals.

3. Simulation model

As mentioned in Section 1, the bubble dynamics in general is well known. We have used the Rayleigh–Plesset equations in the form presented by Barber et al. [24], supplemented with equations for the chemical processes, water vapor balance, and heat balance [25,26] to make simulations of the bubble dynamics in the relevant parameter regime for comparison with experimental results. The instabilities are simulated using the linearized amplitude equation derived by Ref. [5] which is also used by Holzfuss [17]. This includes the boundary layer approximation by Ref. [3] (see also Hao and Prosperetti [4]) that greatly simplifies the original integro-differential equation developed by Prosperetti [2] to treat the non-local nature of the vorticity field induced by the bubble motion due to viscosity. In our simulation only the amplitude of the $n = 2$ shape distortion is computed. The reasons for this choice will be discussed below in Section 5. However, one reason for repeating the simulations presented in Ref. [17] was to see whether the observed period doubling was robust against a change in the model for the radial oscillation.

4. Experimental results

Originally the experiment was designed in order to investigate in detail which type of instability was responsible for bubble extinction. However, when the driving amplitude was set either a little below the onset of emission or just below the upper stability boundary, regularly repeating structures appeared in the Mie scattering signals in the afterbounce region of the bubble oscillation. Here we shall concentrate on the structures seen at the high end of the bubble stability range and especially on those that exhibit period doubling. Let us stress here that even in the regime of the instabilities we are discussing here, the bubble can survive for days.

Even though the signal-to-noise ratio is sufficiently high for the structures to be observed directly, in the following figures we shall display the time series from the three channels averaged over consecutive sets of 4 periods. This has a double advantage. First the averaging removes random noise, secondly it underlines that the periodicity seen is real.